

**Appln No. 10/087,247**

**Amdt date November 25, 2003**

**Reply to Office action of August 25, 2003**

**Amendments to the Claims:**

This listing of claims will replace all prior versions, and listings, of claims in the application:

**Listing of Claims:**

1. (Original) A carbonaceous material comprising:  
a graphite particle having a 002 plane interval  $d_{002}$  of less than 0.337 nm, as measured by the X-ray wide angle diffraction method;  
a complex particle disposed and distributed in the vicinity of the surface of the graphite particle, the complex particle comprising silicon and carbon and having a particle size smaller than that of the graphite particle; and  
an amorphous carbon layer having a 002 plane interval  $d_{002}$  of more than 0.37 nm, as measured by the X-ray wide angle diffraction method, the amorphous carbon layer being a polymer layer and being coated on the graphite particle and the complex particle rendering them bound;  
wherein the complex particle comprises a Si particulate, a conductive carbon material disposed and distributed in the vicinity of the surface of the Si particulate, and a rigid carbon material layer coated on the Si particulate and the conductive carbon material rendering them bound, the Si particulate being composed of a crystalline Si phase.
2. (Original) The carbonaceous material according to claim 1, wherein the crystalline Si phase is deposited with at least

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one phase selected from SiO<sub>2</sub> phases, SiC phases, and SiB<sub>4</sub> phases.

3. (Original) The carbonaceous material according to claim 1, wherein the silicon and the carbon are present in a weight ratio of 0.1:99.9 to 50:50.

4. (Original) The carbonaceous material according to claim 2, wherein the PSiO<sub>2</sub>/PSi ratio is no less than 0.005 and no more than 0.1 and the PSiC/PSi ratio is no less than 0.005 and no more than 0.1, wherein PSi is defined as the diffraction intensity of the plane (111) of the Si phase, PSiO<sub>2</sub> is defined as the diffraction intensity of the plane (111) of the SiO<sub>2</sub> phase, and PSiC is defined as the diffraction intensity of the plane (111) of the SiC phase, measured by the X-ray wide angle diffraction method.

5. (Original) The carbonaceous material according to claim 2, wherein the PSiO<sub>2</sub>/PSi ratio is no less than 0.005 and no more than 0.1, the PSiC/PSi ratio is no less than 0.005 and no more than 0.1, the PSiB/PSiO<sub>2</sub> ratio is no less than 0.1 and no more than 5.0, and a PSiB/PSiC ratio is no less than 0.1 and no more than 5.0, wherein PSi is defined as the diffraction intensity of the plane (111) of the Si phase, PSiO<sub>2</sub> is defined as the diffraction intensity of the plane (111) of the SiO<sub>2</sub> phase, PSiC is defined as the diffraction intensity of the plane (111) of the SiC phase, and PSiB is defined as the diffraction intensity of the plane (104) of the SiB<sub>4</sub> phase, as measured by the X-ray wide angle diffraction method.

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6. (Currently Amended) The carbonaceous material according to Claim 1, wherein the graphite particle has a particle size ranging from ~~2 to 70  $\mu\text{m}$~~ , 2 to 70  $\mu\text{m}$  the complex particle has a particle size of no less than 50 nm and no more than 2  $\mu\text{m}$ , and the amorphous carbon layer has a thickness of no less than 50 nm and no more than 5  $\mu\text{m}$ .

7. (Original) The carbonaceous material according to claim 1, wherein the Si particulate has a particle size of no less than 10 nm and less than 2  $\mu\text{m}$ , the conductive carbon material has a specific resistance of no more than  $10^{-4} \Omega \cdot \text{m}$ , and the rigid carbon layer has a flexibility strength of no less than  $500 \text{ kg/cm}^2$  and a thickness of no less than 10 nm and no more than 1  $\mu\text{m}$ .

8. (Original) The carbonaceous material according to claim 1, wherein the complex particle is present in an amount no less than 1% by weight and no more than 25% by weight.

9. (Original) The carbonaceous material according to claim 1, wherein the amorphous carbon layer is obtained by heat-treating at least one polymer material selected from the group consisting of thermoplastic resins, thermosetting resins, vinyl-based resins, cellulose-based resins, phenol-based resins, coal-based pitch materials, petroleum-based pitch materials, and tar-based materials.

10. (Original) The carbonaceous material according to Claim 9, wherein the mixing weight ratio of Si : graphite : polymer is 0.1:99.8:0.1 to 40:40:20.

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11. (Original) A lithium secondary battery comprising the carbonaceous material according to Claim 1.

12. (Currently Amended) A method of preparing a carbonaceous material comprising the steps of:

calcining a Si particulate composed of a crystalline Si phase in a carbon crucible at ~~1300 to 1400 °C~~ 1300 to 1400 °C to deposit a SiO<sub>2</sub> phase and a SiC phase in the crystalline Si phase;

adding a conductive carbon material to the Si particulate; applying a polymer material coating solution to the Si particulate to provide a complex particle precursor;

calcining the complex particle precursor to render the polymer material coating solution into a rigid carbon layer to provide a complex particle;

adding the complex particle to a graphite particle;

applying a polymer material coating solution to the graphite particle to provide a carbonaceous material precursor; and

calcining the carbonaceous material precursor to render the polymer material coating solution into an amorphous carbon layer to provide a carbonaceous material.

13. (Currently Amended) A method of preparing a carbonaceous material comprising the steps of:

calcining a Si particulate together with a B<sub>2</sub>O<sub>3</sub> powder in a carbon crucible at ~~1300 to 1400 °C~~ 1300 to 1400 °C to deposit SiO<sub>2</sub>, SiC, and SiB<sub>4</sub> phases in a crystalline Si phase;

adding a conductive carbon material to the Si particulate;

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applying a polymer material coating solution to the Si particulate to provide a complex particle precursor;

calcining the complex particle precursor to render the polymer material coating solution into a rigid carbon layer to provide a complex particle;

adding the complex particle to a graphite particle;  
applying a polymer material coating solution to the graphite particle to provide a carbonaceous material precursor; and  
calcining the carbonaceous material precursor to render the polymer material coating solution into an amorphous carbon layer to provide a carbonaceous material.